PATENT COOPERATION TREATY

PCT

REC'D 13 JUL 2000

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

16

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 2996108	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/SE99/00343	International filing date *(day/month/year)* 08.03.1999	Priority date *(day/month/year)* 17.03.1998
International Patent Classification (IPC) or national classification and IPC7 B01D 53/56, B01D 53/86, F23J 15/08		
Applicant HEED, Björn		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 4 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of _____ sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
12.10.1999	05.07.2000
Name and mailing address of the IPEA/SE Patent- och registreringsverket Box 5055 S-102 42 STOCKHOLM Facsimile No. 08-667 72 88	Telex 17978 PATOREG-S
Authorized officer Marianne Bratsberg/MP Telephone No. 08-782 25 00	

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

[redacted]national application No.
PCT/SE99/00343

I. Basis of the report

1. This report has been drawn on the basis of *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.)*:

☒ the international application as originally filed.

☐ the description, pages _____ , as originally filed,
pages _____ , filed with the demand,
pages _____ , filed with the letter of _____ ,
pages _____ , filed with the letter of _____ .

☐ the claims, Nos. _____ , as originally filed,
Nos. _____ , as amended under Article 19,
Nos. _____ , filed with the demand,
Nos. _____ , filed with the letter of _____ ,
Nos. _____ , filed with the letter of _____ .

☐ the drawings, ~~sheets~~/fig _____ , as originally filed,
~~sheets~~/fig _____ , filed with the demand
~~sheets~~/fig _____ , filed with the letter of _____ ,
~~sheets~~/fig _____ , filed with the letter of _____ .

2. The amendments have resulted in the cancellation of:

☐ the description, pages _____
☐ the claims, Nos. _____
☐ the drawings, ~~sheets~~/fig _____

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the supplemental Box (Rule 70.2(c)).

4. Additional observations, if necessary:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

[redacted]national application No.
PCT/SE99/00343

V. Resoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims	1-5	YES
	Claims		NO
Inventive step (IS)	Claims		YES
	Claims	1-5	NO
Industrial applicability (IA)	Claims	1-5	YES
	Claims		NO

2. Citations and explanations

The invention relates to equipment for purification of gases using heat-exchanging matrices to heat the gases to oxidation or decomposition temperature. At least one of the heat exchanging matrices is catalytically active in promoting reduction of NOx.

A corrected International Search Report has been established on 17th of April 2000, citing the following documents:

D1: US 5262131
D2: US 4741690
D3: US 5178101
D4: WO 9307954

The closest prior art is regarded to be D1, which describes a catalytic regenerative thermal reactor. The reactor includes a bed of regenerative heat transfer material to preheat the incoming contaminated air (i.e. a heat-exchanging matrix), an oxidation catalyst distributed in or on the heat transfer material and means for heating the catalyst. (See column 4, line 32-39)
In one of the embodiments the reactor is described to consist of several stacked layers or beds of silica or gravel with two catalyst beds placed in between these. (See abstract)

The invention according to claim 1 differs from D1 in that the purpose of the catalyst is to reduce nitrogen oxides (NOx). In D1 the catalyst is used for oxidation of volatile organic compounds (VOC). However, it is considered to be obvious for a person skilled in the art to replace the catalyst for oxidising VOC described in D1 with a catalyst for reduction of NOx when excessive NOx is regarded to be the aim of the purification operation.

.../...

Supplemental Box
(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of: V.

D2 describes an arrangement, which only differs from the invention according to claim 1 in that there is no catalyst present in the matrix (see abstract). Even though D2 describes a number of disadvantages in using catalysts it is considered to be obvious for a person skilled in the art to incorporate a catalyst for reducing NOx emissions since one of the main object in D2 is to reduce NOx (see column 1, line 64 - column 2, line 2), c.f. D1.

The invention according to claim 2 does not include any characteristics, which not already have been discussed above or can be found in D1.

Claim 3 further describes the invention to include means for injecting reducing agents in the incoming gas flow. This measure is common in the art of reducing NOx in exhaust gases and is for example described in D3 (see claim 1). Claims 4 and 5 regard the control of the injection of reducing agents. It is considered to be obvious for a person skilled in the art to control the dosage and timing of the injections in an appropriate way to make sure that the reducing agent will work efficiently and be consumed. This reasoning can also be found in D3 (column 1, line 51-66). Furthermore, claims 4 and 5 are characterised by a wish to obtain a certain goal and not by distinctive, technical features. Hence, the invention according to claims 3-5 lacks an inventive step.

Hence, the invention according to claims 1-5 is novel but regarded to lack an inventive step.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SE 99/00343

A. CLASSIFICATION OF SUBJECT MATTER

IPC7: B01D 53/56, B01D 53/86, F23J 15/08
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: B01D, F23J, F23G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI,EPODOC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5262131 A (BAYER ET AL), 16 November 1993 (16.11.93), column 2, line 29 - column 3, line 58; column 1, line 43 - column 2, line 18, figure 1, claim 1, abstract --	1-5
X	US 4741690 A (BJÖRN HEED), 3 May 1988 (03.05.88), column 1, line 60 - column 2, line 22; column 2, line 59 - column 3, line 41, figure 1, abstract --	1-5
A	US 5178101 A (RONALD D. BELL), 12 January 1993 (12.01.93), column 1, line 51 - line 66; column 5, line 41 - column 7, line 65, figure 1, claim 1, abstract --	1-5

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:
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"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
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Date of the actual completion of the international search	Date of mailing of the international search report
17 April 2000	28 -04- 2000
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer Marianne Bratsberg/ELY Telephone No. +46 8 782 25 00

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SE 99/00343

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 9307954 A1 (HEED, BJÖRN), 29 April 1993 (29.04.93), column 1, line 5 - column 2, line 13	1-5

CORRECTED

PCT

RLD INTELLECTUAL PROPERTY ORGANIZAT
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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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B01D 53/56, 53/86, F23J 15/08

A1

(11) International Publication Number: WO 99/47245

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(21) International Application Number: PCT/SE99/00343

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(30) Priority Data:
9800866-7 17 March 1998 (17.03.98) SE

(71)(72) Applicant and Inventor: HEED, Björn [SE/SE]; Utlandagatan 19, S-412 61 Göteborg (SE).

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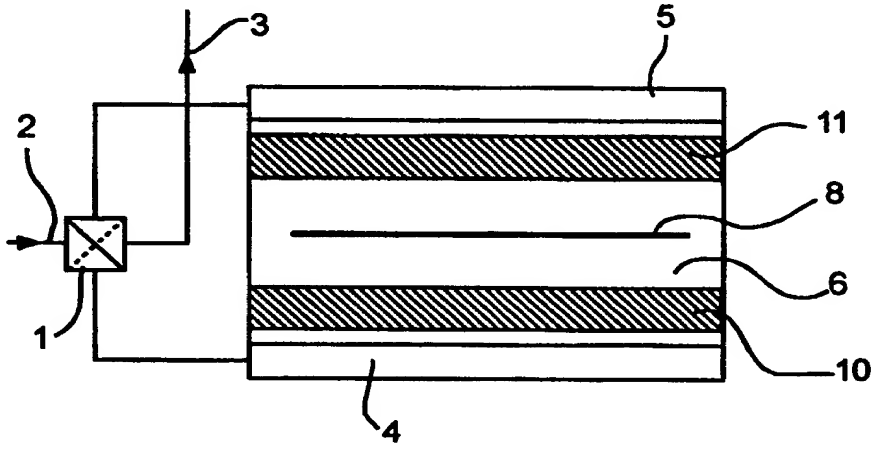
(81) Designated States: AL, AM, AT, AT (Utility model), AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, CZ (Utility model), DE, DE (Utility model), DK, DK (Utility model), EE, EE (Utility model), ES, FI, FI (Utility model), GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (Utility model), SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published
With international search report.

(54) Title: POLLUTION CONTROL

(57) Abstract

Device for pollution control where a polluted stream of air or gas is purified from both oxidisable material and nitrogen oxides simultaneously by a combination of regenerative high temperature treatment and catalytic treatment.



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POLLUTION CONTROL

Polluted air and other gases can be purified by heat treatment to such temperatures that the pollutants are oxidised or decomposed. US patent 4 267 152 and US patent 4 741 690 describes such processes where the polluted gases are fed through regenerative devices where the heating of a gas is immediately followed by cooling and recovery of the heat content of the gas. In this way heat treatment of the gas to a high temperature can be made in an economical way without a high expenditure of energy.

The incoming raw gas is in these processes successively heated to the high temperature by means of contact with a matrix of solid material capable of heat transfer to the gas. In the solid matrix there is a temperature gradient so that the gas is first successively heated to a maximum temperature. After attaining its maximum temperature the gas is then cooled in an analogous manner by means of contact with a solid matrix of successively lower temperature. In US patent 4 287 152 the heating and the cooling matrices are separated from each other but used alternatively for heating and cooling purposes according to an alternating direction of gas flow through the matrices. The different matrices are alternatively used for heating and cooling of the gas. In US patent 4 741 690 there is only one continuos matrix through which the gas flow is being fed. The temperature profile in this matrix is such however that when the gas passes through it is first successively heated to a maximum temperature and then successively cooled.

In both cases the operation is regenerative and the gas is fed in alternating directions through the equipment and is successively first heated to a maximum temperature and then cooled. The maximum temperature employed is such that it is at or above the temperature that is necessary for the intended oxidation or decomposition reaction to take place.

Processes and equipment like these are extensively used for the purification of ventilation air from painting and printing processes. It can also be used for the purification of exhaust gases from internal combustion engines. In this and other cases nitrogen oxides are a part of the pollution problem.

For diesel engines the concentration of nitrogen oxides in the exhaust can reach several thousands of ppm. Good reduction of these nitrogen oxides can be accomplished if the exhaust is first mixed with a corresponding amount of ammonia, urea or other amine compound before it is heat treated in the above described manner. During the heating the mixture will pass through the relevant temperature window for a selective non catalytic reduction (SNCR) reaction where the nitrogen oxides and the amines are transformed to elemental and harmless nitrogen.

Diesel engine exhaust for example can thus be purified in the above described process first during the heating phase when nitrogen oxides are removed and then at the high temperature when soot, aldehydes and other organic matter is oxidised. This way of operation of the equipment is described in European patent number EPC 609 288.

Experience has shown that when the original concentration of nitrogen oxides is more than 1000 ppm a good reduction can be achieved and the concentration nitrogen oxides brought down to a couple of hundred ppm. However when the original concentration already is in this low region as for example in the exhaust from lean burn natural gas engines reduction is very poor.

The present invention constitutes a way of improving this situation so that the concentration of nitrogen oxides in the outgoing treated gas can be brought down significantly below 50 ppm. The improvement is accomplished by the incorporation of catalytically active zones into the heat transfer matrices of earlier designs.

An embodiment of the invention is described in figure 1 and another embodiment of the invention is described in figure 2. In both figures 1 is a valve mechanism for the direction of air (gas) in alternating directions through the equipment.2 and 3 are connecting ducts for incoming raw and outgoing cleaned gas respectively. 4 and 5 are wind boxes for distribution and collection of air (gas) that goes through the heat transfer matrix 6. In the design shown in figure 2 this matrix is divided into two parts 6 and 6' surrounding a combustion chamber 7 which is absent in the design shown in figure 2 where the heat exchanging matrix stretches all the way between the two wind boxes 4 and 5. Both designs comprises means for heating: in figure 1 in the form of electric heaters 8 and in figure 2 in the form of a burner 9. Both designs also incorporate catalytically active zones 10 and 11 within the heat exchanging matrices. In the design of figure 1 the temperature is high in the centre of the heat exchanging matrix 6 and gradually decreases towards top and bottom. In the design in figure 2 the temperature

is high in the combustion chamber 7 and the upper parts of the heat exchanging matrixes 6 and 6′ and the temperature gradually decreases towards the bottom of the heat exchanging matrices. By regenerative heat exchange and regular switching of the direction of flow through the equipment these temperature patterns can be generally maintained without excessive heat demands being put on the heating means 8 (figure 1) and 9 (figure 2). When the oxidation of pollutants in the gas stream produces enough energy they can be switched off altogether.

In operation raw polluted gas is first mixed with ammonia, urea, or other compound able to act as a selective nitrogen oxides reducing agent. The mixture is then introduced into a cold end of a heat exchanger matrix and by passing through it is successively heated to oxidation or decomposition temperature which is attained in the inner part of the equipment i.e. the hot centre of the matrix in fig 1 or the uppermost parts of the matrices 6 and 6′ and the combustion chamber 7 in fig 2. Before reaching this high temperature however the mixture is passed through the catalytically active zone of the heat exchanger matrix 6 or 6′. Here nitrogen oxides react with the mixed in reducing agent and are thus removed. The catalytic zone is placed in the heat exchanging matrix in such a way that the temperature conditions there are favourable for this reduction and a selective catalytic reaction (SCR) takes place. The reaction takes place at a substantially lower temperature than an SNCR reaction and this together with the use of catalyst makes a more thorough reduction possible as compared with an operation according to European patent EPC 609 288 and lower levels of concentration of nitrogen oxides are obtainable. When the inlet concentration of

nitrogen oxides is low the difference becomes significant.

After the SCR reaction the gas mixture is further heated and as in European patent EPC 609 288 other pollutants as well as any remaining surplus of reduction agents are destroyed in the hot inner part.

In operation of regenerative equipment like this the direction of gas through the equipment is reversed at regular intervals. Unless special precautions are taken, at every change in flow direction some untreated gas mixture is being "short circuited" or carried over to the outlet. It is then advantageous to interrupt the supply of reducing agent for a short period before such changes in flow direction. Unnecessary emissions of reducing agent is then avoided.

An important aspect of the invention is that catalyst can be applied in such a way that the reducing activity of the catalyst is retained for a considerable time after the supply of reducing agent has been interrupted. The overall reduction efficiency of the equipment thus is not disadvantageously affected by such interruptions in the supply of reducing agent. This effect can be so pronounced that the equipment can be modified to comprise only one zone of catalytically active material. This zone is then activated by the supply of reducing agent when this part of the equipment is used as inlet end for the gas stream. When the gas stream is reversed and the active zone is at the outlet end of the equipment the supply of reducing agent is cut off and the reduction of nitrogen oxides takes place in this zone after the high temperature treatment. Figure 3 shows such a design with only one catalytically active zone 10.

The invention has been described above in the conjunction with regenerative equipment using either one heat exchanging matrix or two different matrices surrounding a combustion chamber. There also are designs using three or more heat exchanging matrices surrounding a common combustion chamber. In some designs the direction of flow through the heat exchanging matrix is changed only gradually in the heat exchanging matrix so that different parts of the same matrix have flow in different directions. This is obtained for instance by rotation of a matrix versus fixed inlet and outlet ports or by the use of a rotating valve system working together with a fixed matrix. Together all these different designs are often called regenerative thermal oxidisers (RTOs). In all the different designs the heat exchanging matrix material is subject to a gas flow that is reversed at regular intervals and the incoming gas is successively heated to a high temperature where oxidation and decomposition takes place. The invention is applicable in all these cases. The design shown in figures 1 and 3 have the advantage that the equipment is compact and can be made comparatively small which very often is an important aspect when used in various conjunctions together with internal combustion engines.

Catalytically active zones can be incorporated in at least one of the matrices and be activated by regular supply of reducing agent. As described above this supply does not have to be continuous. In some cases the raw gas may already contain such reducing agents or the catalyst or combination of catalyst and raw gas be such that no such supply is necessary.

CLAIMS

1. Equipment for the purification of gases employing one or several heat exchanging matrices where the gas in a regenerative process is heated to oxidation or decomposition temperature, characterised by that at least one of the heat exchanging matrices comprises a zone that is catalytically active in promoting reduction of nitrogen oxides.

2. Equipment for the purification of gases employing a single heat exchanging matrix where the gas is heated in a regenerative process to oxidation or decomposition temperature, characterised by that the heat exchanging matrix comprises two zones that are catalytically active and situated on each side of the hot centre zone of the matrix.

3. Equipment according to claims 1 or 2, characterised by that it comprises means for the supply of agents to the incoming gas flow that reduce nitrogen oxides.

4. Equipment according to claim 3, characterised by that the supply of reducing agent is interrupted for a short while in connection with change of direction of gas flow through the equipment.

5. Equipment according to claim 1, characterised by that supply of reducing agent is maintained only when the gas to be treated goes through the equipment in such a way that it passes a zone that is catalytically active before it reaches temperatures that are so high that oxidation or decomposition occurs.

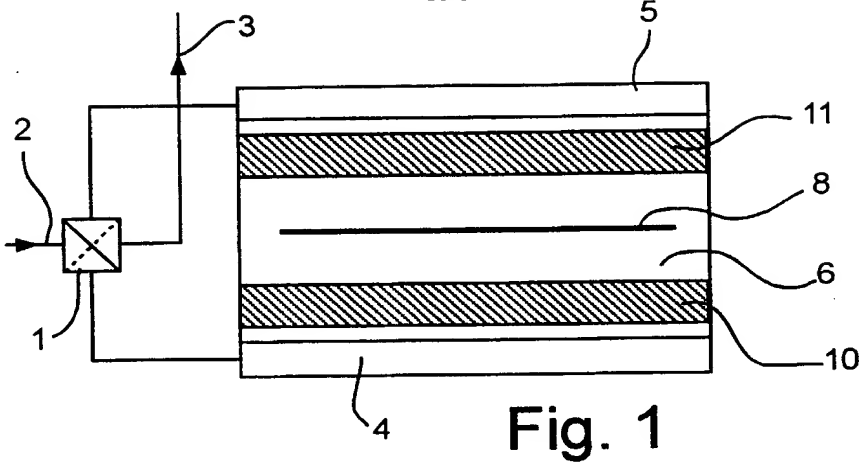

Fig. 1

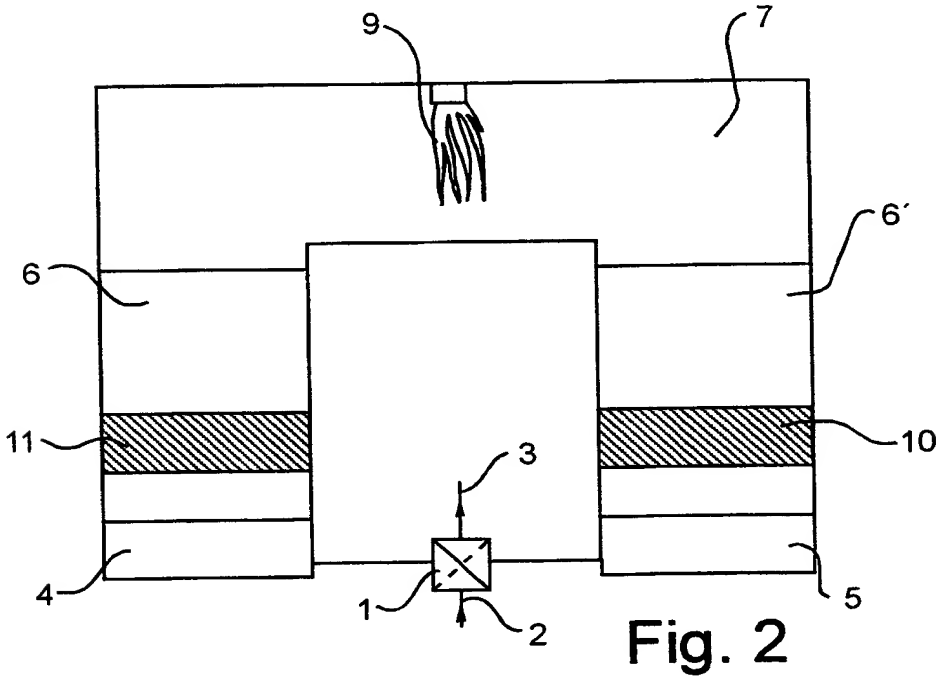

Fig. 2

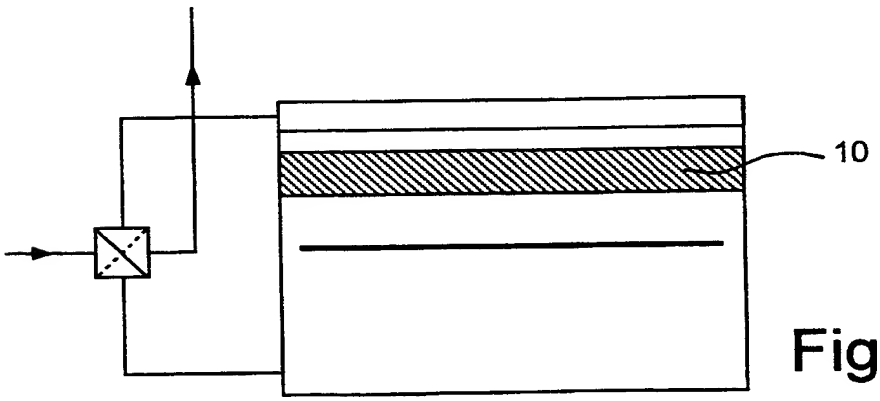

Fig. 3

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SE 99/00343

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B01D 53/56, B01D 53/86, F23J 15/08
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: B01D, F23J, F23G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI,EPODOC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5178101 A (RONALD D.BELL), 12 January 1993 (12.01.93), figure 1, claim 1, abstract ---	1-5
A	US 4741690 A (BJÖRN HEED), 3 May 1988 (03.05.88) ---	1-5
A	WO 9307954 A1 (HEED, BJÖRN), 29 April 1993 (29.04.93) --- --------	1-5

☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
19 May 1999	15-06-1999
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer Britt-Marie Lundell Telephone No. +46 8 782 25 00

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

03/05/99

International application No.
PCT/SE 99/00343

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5178101 A	12/01/93	US 5224334 A	06/07/93
		WO 9317802 A	16/09/93
US 4741690 A	03/05/88	AT 41052 T	15/03/89
		CA 1249213 A	24/01/89
		DE 3590307 T	04/06/87
		EP 0218590 A,B	22/04/87
		SE 0218590 T3	
		JP 7033905 B	12/04/95
		JP 61502484 T	30/10/86
		SE 441623 B,C	21/10/85
		SE 8403330 D	00/00/00
		WO 8600389 A	16/01/86
WO 9307954 A1	29/04/93	AT 135254 T	15/03/96
		AU 2759192 A	21/05/93
		CA 2121238 A	29/04/93
		DE 69209084 D,T	22/08/96
		EP 0609288 A,B	10/08/94
		ES 2084385 T	01/05/96
		JP 7500280 T	12/01/95
		SE 468156 B,C	16/11/92
		SE 9103008 A	16/11/92